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Using Chlorine Dioxide for Trihalomethane Control

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16. ABSTRACT

Because of the November 1979 amendment to the National Interim Primary Drinking Water Regulations, many utilities were faced with changing their disinfection practices to comply with the 0.10-mg/L maximum contaminant level for trihalomethanes (THMs). After considering various options, one utility, the Evansville (Ind.) Water and Sewer Utility, chose chlorine dioxide disinfection as the most feasible method for controlling THMs. The performance of a 100-gpm (6.3-L/s) pilot plant that incorporated chlorine dioxide was so effective for reducing THMs that it has been used in Evansville's full-scale plant since 1983.

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JOURNAL ARTICLE

Using Chlorine Dioxide for Trihalomethane Control

Benjamin W. Lykins Jr. and Mark H. Griese

Because of the November 1979 amendment to the National Interim Primary Drinking Water Regulations, many utilities were faced with changing their disinfection practices to comply with the 0.10-mg/L maximum contaminant level for trihalomethanes (THMs). After considering various options, one utility, the Evansville (Ind.) Water and Sewer Utility, chose chlorine dioxide disinfection as the most feasible method for controlling THMs. The performance of a 100-gpm (6.3-L/s) pilot plant that incorporated chlorine dioxide treatment was compared with that of a full-scale plant that used chlorination. Chlorine dioxide was so effective for reducing THMs that it has been used in Evansville's full-scale plant since 1983.

The use of chlorine in water treatment, an accepted practice for many years, has been one of the major factors in reducing waterborne disease. The discovery of trihalomethanes (THMs) and their potential health effects, however, raised questions about the safety of chlorination, which, while protecting drinking water against infectious disease, may also produce carcinogenic organic by-products.[1] This concern produced an amendment to the National Interim Primary Drinking Water Regulations to control the concentration of THMs in drinking water.

In order to comply with the THM regulation, the Evansville (Ind.) Water and Sewer Utility had the same options as many other utilities: use a disinfectant other than chlorine (Cl_2), change the point of chlorination, improve existing clarification for THM precursor reduction, or use an adsorbent (powdered activated carbon [PAC] or granular activated carbon [GAC]) for THM or precursor removal.[2] Staff at the Evansville utility selected an alternative disinfectant—chlorine dioxide (ClO_2)—as the most viable option, but not without reservations. There were concerns about operation and maintenance, inorganic products such as chlorite (ClO_2^-) and chlorate (ClO_3^-), increased disinfection costs, and possible health effects.

To address some of these concerns, the Evansville utility, with the US Environmental Protection Agency (USEPA), initiated research to evaluate the use of ClO_2. This evaluation was conducted in a 100-gpm (6.3-L/s) pilot plant constructed adjacent to Evansville's 60-mgd (227-ML/d) full-scale plant, with parallel performances for the two plants being compared. The effectiveness of ClO_2 resulted in the utility changing its pre-disinfectant from Cl_2 to ClO_2. This article presents the results of the pilot-plant evaluation and describes the subsequent use of ClO_2 in the full-scale plant.

Full-scale treatment plant

The Evansville filtration plant, located on the Ohio River about 1 mi (1.6 km) upstream from the city of Evansville, was used as the control for the pilot-plant research study. The plant consists of two separate treatment systems, each having a hydraulic capacity of about 30 mgd (113.5 ML/d). One system, designated the south plant, was the control.

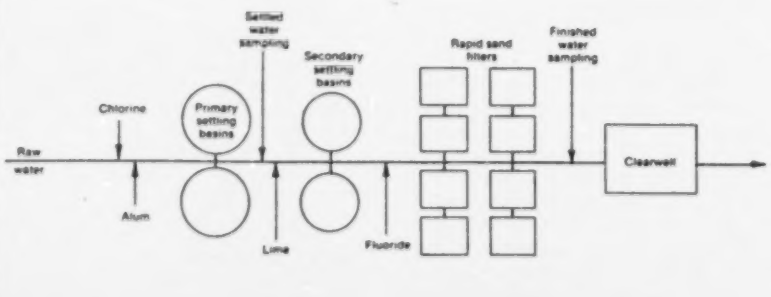

Figure 1. Flow diagram of south system of full-scale plant

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TABLE 1

Experimental modes of operation used during optimization phase of pilot testing

Run	Chlorine	Chlorine Dioxide	Alum	Polymer	Mix	Settle	Lime	Filter	Carbon	Chlorine	Chlorine Dioxide
1	X		X	X	X	X	X	X		X	
2	X		X	X	X	X	X	X	X	X	
3			X	X	X	X	X	X	X		X
4		X	X	X	X	X	X	X	X		X

The south plant consists of two primary settling basins, two secondary settling basins, and eight rapid sand filters (Figure 1). Each primary settling basin is 130 ft (39.6 m) in diameter and 17.5 ft (5.3 m) deep, with a capacity of approximately 1.8 mil gal (6.8 ML). The secondary settling basins are each 90 ft (27.4 m) in diameter with a 15-ft (4.5-m) sidewall depth and a capacity of 0.725 mil gal (2.7 ML).

Average concentrations of 6 and 28 mg/L of chlorine and alum, respectively, were added prior to primary settling. These dosages were altered according to the demand and turbidity of the raw water. A free Cl_2 residual of 1.5–2.0 mg/L was maintained after sand filtration, and, in order to maintain this residual, postchlorination occurred before the water passed into a common clearwell. Approximately 12 mg lime/L for pH adjustment to 8 was added after primary settling, and about 1.0 mg fluoride/L was added immediately before the rapid sand filters.

Pilot plant

A 100-gpm (6.3-L/s) unit* was housed on-site for studying treatment and operational options at Evansville without disrupting the normal water supply procedure. This pilot plant consisted of a rapid mix chamber, a mechanical flocculator, and a settling tube chamber for clarification (Figure 2). Clarified water then passed through a mixed-media filter bed to an underdrain system, where it was pumped to two parallel pressure granular activated carbon contactors or directly to a combination clearwell and backwash tank.

Average alum and anionic polymer dosages of 12 and 0.8 mg/L, respectively, were used for turbidity removal. Although not used in the full-scale plant, a polymer was necessary in the pilot plant for effective turbidity control because of the short detention time (37 min) of the water. About 6 mg lime/L for pH adjustment to 8 was added to the settling tube effluent. Disinfection with Cl_2 was achieved by using a 3-percent sodium hypochlorite (NaOCl) solution, whereas ClO_2 was produced by a generator.† Using this generator, NaOCl was reacted with sodium chlorite ($NaClO_2$) after the pH of the reaction had been adjusted to 3.0 ± 0.5 with hydrochloric acid (HCl).

Two sequential phases were used to evaluate pilot-plant operation: optimization and long-term.

Optimization phase

The optimization phase consisted of a control study and three experimental modes of operation. For the control study, the pilot plant was operated in a mode similar to that of the full-scale plant, namely, prechlorination (before coagulation and settling) and postchlorination (after mixed-media filtration). The three experimental modes were evaluated to determine which treatment method would be used during the long-term operating phase (Table 1).

The performance of the pilot plant was compared with that of the full-scale plant to determine whether the full-scale plant could be used as a control for subsequent pilot-plant studies. As shown in Table 2, the average compositions of the two plant effluents were comparable. The data for total instantaneous THMs and total organic carbon (TOC) were subjected to the *t*-test at a 95-percent confidence level. No significant difference between the two plants was noted for either parameter.

During another study in the pilot plant, predisinfection was not used so that the effectiveness of coagulation-settling and filtration could be determined before the performance of ClO_2 was evaluated. This study showed that

- the low instantaneous THM concentration (1.2 μg/L) in the raw water was not reduced during treatment,
- the raw water TOC concentration was reduced by 40 percent from 4.2 mg/L to 2.5 mg/L,
- the raw water turbidity was reduced 98 percent from 20.6 to 0.3 ntu, and
- the raw water total coliforms and heterotrophic plate counts were reduced from 5400 to 3800/100 mL and 290 to 45/mL, respectively.

During this study, samples were also stored with a Cl_2 or ClO_2 residual for three days to simulate Evansville's distribution time. Table 3 shows the differences in average THM concentrations that could be expected if predisinfectants were not used and the plant effluent was disinfected with either Cl_2 or ClO_2 (141 μg/L versus 1.4 μg/L total THMs, respectively).

During the final optimization run, ClO_2 was added to the raw water in the rapid mix chamber. As expected, the THM concentration did not increase through the pilot plant. Adequate disinfection was accomplished, with the total coliforms being reduced from 6300/100 mL to below detection and the heterotrophic plate count being reduced from 6200 to 16/mL. This disinfection occurred even though the ClO_2 residual decreased dramatically through the pilot treatment plant (4 to 0.3 mg/L).

Adequate procedures were not available during the pilot-plant study to determine the concentrations of ClO_3^- and chloride (Cl^-). However, several methods were considered,[3-7] and analyses were

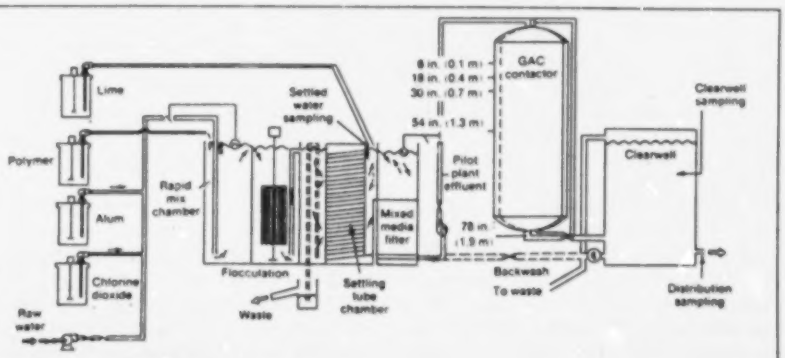

Figure 2. Flow diagram of pilot plant

*Neptune Microfloc, St. Paul, Minn.
†Olin Corporation, Overland Park, Kan.

run using the LaMotte-Palin modified titration procedure[8,9] and the chlorophenol red titration method.[10]* As the ClO_2 concentration decreased, the ClO_2^- concentration generally increased. Since this increase was not proportional to the decrease in ClO_2, other species, such as Cl^-, were probably formed after the ClO_2 demand had been satisfied.

Long-term evaluation

In the long-term study with pre-ClO_2 disinfection, four runs were completed that consisted of continuous pilot-plant operation for approximately three months each. The pilot plant was evaluated during each season of the year to determine the extent of seasonal effects. Figure 3 shows the effect of the seasons on the THM concentration in the effluent of the full-scale plant, probably the result of temperature and precursor changes.

Samples of raw water, pilot-plant effluent, and full-scale plant effluent were stored for three days at ambient temperature and a pH of 8 with a free Cl_2 residual to simulate the residence time at the farthest point in Evansville's distribution system. This was done to compare a system using pre- and post-chlorination (full-scale plant) with a similar one using pre-ClO_2 and post-chlorination (pilot plant). Because of the difficulty in maintaining a ClO_2 residual, postchlorination is required to maintain a disinfectant residual in the distribution system.

Figure 4 illustrates the reduction in the concentration of terminal THMs that was noticed when raw water was disinfected with ClO_2. Pre-ClO_2 disinfection with delayed chlorination permitted coagulation-settling-filtration and oxidation to remove THM precursors, thereby reducing the amount of THMs formed during posttreatment chlorination. A comparison of the average terminal THM concentrations for the two disinfectant modes showed a reduction of approximately 60 percent when ClO_2 was used. Although THM precursors were reduced by using pre-ClO_2, no difference was noted between the average TOC concentrations, regardless of the type of predisinfectant used (Figure 5).

Because of the THM reductions resulting from pre-ClO_2 disinfection, an additional study was done to reaffirm the feasibility and benefits of using ClO_2 in Evansville's full-scale plant. Although ClO_2 could be used as a predisinfectant, chlorine was required as a postdisinfectant because of the size of Evansville's distribution system and the need to maintain a sufficient disinfectant residual throughout the system. Data generated from previous tests indicated that maintaining a ClO_2 residual over an extended period of time is difficult.

*Olin Corporation, Overland Park, Kan.

TABLE 2

*Comparison of pilot- and full-scale plant effluent samples**

Parameter	Raw Water	Pilot-Plant Effluent†	Full-Scale Plant Effluent
Instantaneous THM—*μg/L*	1.9	33.6	36.9
TOC—*mg/L*	2.9	2.1	1.8
1,2-Dichloroethane—*μg/L*	<0.1	<0.1	<0.1
Tetrachloroethylene—*μg/L*	<0.1	<0.1	<0.1
Arsenic—*μg/L*	2.6	<0.1	<0.1
Barium—*mg/L*	0.14	0.10	0.12
Cadmium—*mg/L*	<0.002	<0.002	<0.002
Chromium—*mg/L*	0.014	<0.002	<0.002
Lead—*mg/L*	<0.01	<0.01	<0.01
Mercury—*μg/L*	<0.2	<0.2	<0.2
Selenium—*μg/L*	<1.0	<1.0	<1.0
Silver—*mg/L*	<0.001	<0.001	<0.001
Turbidity—*ntu*	77.0	0.3	0.2
Chlorine—*mg/L*	ND‡	1.5	1.9
Median pH	7.5	7.5	7.4
Temperature—*°F (°C)*	55.9 (13.3)		58.2 (14.6)
Iron—*mg/L*	1.3	0.02	0.02
Manganese—*mg/L*	0.7	<0.1	<0.1
Coliforms—*number/100 mL*	13 000	ND	ND
Heterotrophic plate count—*number/mL*	1 400	ND	ND

*Average of 10 samples over a two-week period for all parameters, except 6 samples for TOC and 2 for heavy metals
†Operational mode: run 1, Table 1
‡ND—not detected

TABLE 3

Simulated production of THMs in distribution system—no predisinfection

Parameter	Pilot Plant Effluent	
	After Cl_2 Disinfection	After ClO_2 Disinfection
Instantaneous THM*—*μg/L*		
Three-day stored THM—*μg/L*	141	1.4
Median pH	7.9	7.8
Temperature—*°F (°C)*	80.6 (27)	80.6 (27)
Chlorine residual—*mg/L*†	2.5	
Chlorine dioxide residual—*mg/L*†		1.9

*Instantaneous THM of raw water—1.2 μg/L
†Residual after 3 days; dosage—3–7 mg/L

TABLE 4

Comparison of filter effluents of north and south systems of Evansville's full-scale plant

	No Predisinfection*	Chlorine Dioxide Added to South System Only†
Average THMs of north system—*μg/L*	29.1	37.3
Average THMs of south system—*μg/L*	28.9	25.5

*3/26/84-3/30/84
†1.4 mg ClO_2/L; 4/12/84-5/3/84; no predisinfection of north system

Chlorine dioxide was introduced into the raw water in sufficient quantities to maintain a slight residual through mixed-media filtration (average addition—1.1 mg/L; residual after filtration—0.1 mg/L). Chlorine was introduced into the water before it entered the pilot-plant clearwell. The average residual concentration of Cl_2 in the clearwell was 2.1 mg/L, which was representative of that used in Evansville's normal treatment process.

As in previous tests, routine sampling for turbidity, pH, temperature, Cl_2 and ClO_2 residuals, total coliforms, and heterotrophic plate counts was performed at both plants. In addition, the raw water, pilot-plant filter effluent and clearwell, and the full-scale plant effluent were analyzed for instantaneous THMs. The instantaneous THM data again showed a reduction in THMs when pre-ClO_2 disinfection was implemented. No difference in the bacteriological quality of the chlorinated effluents of the plants was noticed.

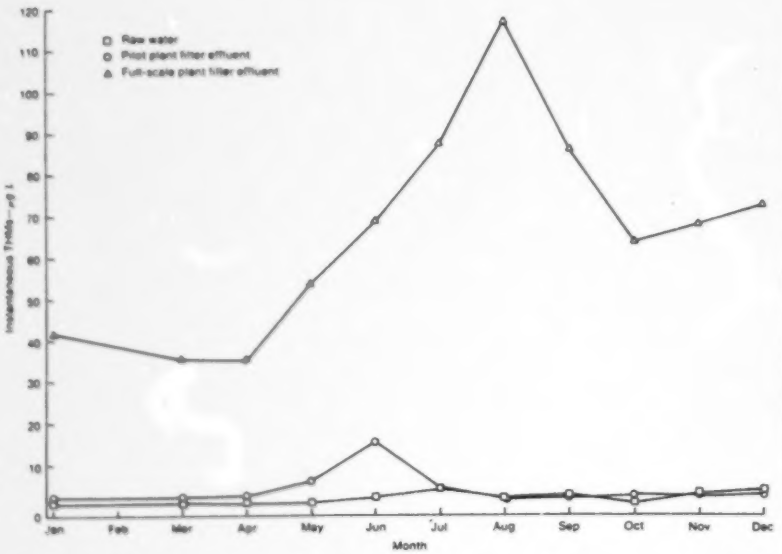

Figure 3. Effect of the seasons on average instantaneous THM concentrations

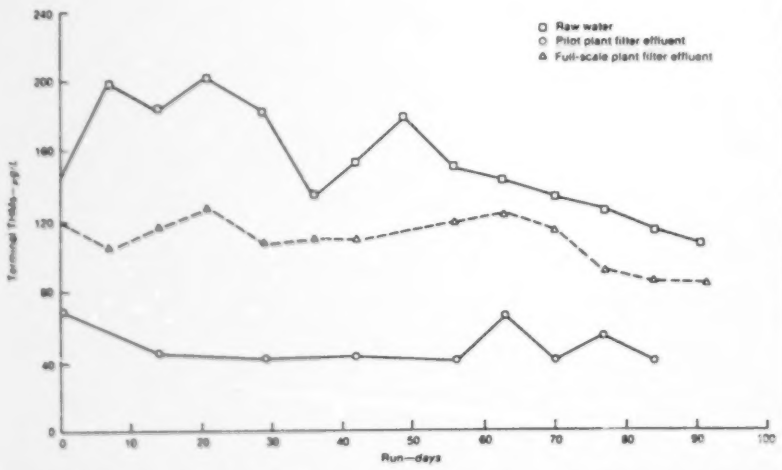

Figure 4. Average terminal THM concentrations in effluents of full-scale and pilot plants

Although ClO_2 was shown to reduce THM concentrations, questions arose concerning other potentially harmful chemicals that might be formed by its use. Gas chromotography–mass spectrometry, however, did not identify any organic by-products attributable to the use of ClO_2. This observation has been substantiated by another disinfection study at Jefferson Parish, La.[11]

The concentrations of ClO_2 and its reduction product ClO_2^- varied during treatment. With the generator used in this study, a stoichiometric reaction was not possible. This generator produced an average percentage yield of 64, 30, and 6 for ClO_2, ClO_2^-, and free Cl_2. Averages of about 0.7 mg ClO_2^-/L and 0.2 mg Cl_2/L were introduced when an average dose of 1.6 mg ClO_2/L was applied to the raw water. After filtration, the concentrations of ClO_2 and ClO_2^- were in the range of 0.3–0.5 and 1.5–2.0 mg/L, respectively, depending on the raw water dosage.

In the previous test, total coliforms and heterotrophic plate counts were adequately reduced using pre-ClO_2 disinfection. However, the length of the long-term evaluation and the associated seasonal variations in the raw water bacterial counts resulted in somewhat higher counts in the finished water. The average total coliform counts of the raw water averaged 1400 to 31 500/100 mL for the four long-term runs. With pre-ClO_2 disinfection, the coliforms (after filtration) were in the range of below detection to 7/100 mL. The average raw water heterotrophic plate count ranged from 1500 to 6300/mL. After filtration, the average range was 1–80/mL. No coliforms or plate counts were detected after postchlorination.

Some bacteriological speciation was attempted. By using an R-2A medium,[12] pigmented bacteria could be isolated. At times, no significant pigmented bacterial populations were detected when sufficient ClO_2 had been applied to the raw water to maintain a residual of 0.3–0.5 mg/L after filtration. At other times, a few pigmented bacteria were seen. Identification of the pigmented bacteria was difficult and not successful.

Full-scale implementation

Obtaining USEPA and state approval. Based on data generated by the pilot-plant evaluation and on the characteristics of Evansville's raw water source, utility officials concluded that the maximum contaminant level (MCL) for total trihalomethanes (TTHMs) was likely to be exceeded if treatment was not altered. This belief was confirmed by preliminary USEPA sampling results. After studying the alternatives available for reducing THMs and concluding that changing the primary disinfectant from Cl_2 to ClO_2 would provide the most effective control at the least cost, a request was made to the USEPA Region V and the Indiana State Board of Health for approval of this treatment modification. After a visit by officials from both agencies and their review of the facilities and the information gathered from the research project, the utility was given approval for the proposed change. In an effort to ensure that water quality would not be adversely affected by this modification, approval was subject to the following conditions:

- Monitoring would be initiated to acquire baseline data for heterotrophic plate counts at several locations in the treatment plant and distribution system. This monitoring would be carried out for six months prior to the treatment change and 12 months following the change and would indicate any deterioration in the bacteriological quality of the water.
- Daily monitoring of total oxidant levels (the sum of ClO_2, ClO_2^-, and ClO_3^-) would be initiated at the time of the treatment change and would be reported to the state board of health on a monthly basis.

- The system would be operated to maintain a total residual oxidant concentration of <1.0 mg/L in the distribution system.
- Any degradation of water quality would immediately be reported to the USEPA and the Indiana State Board of Health to permit reassessment of the treatment change.

Generating chlorine dioxide. While the baseline data were being collected, construction was begun on a separate building to house the ClO_2 generation equipment. One portion of the building contained the ClO_2 generator, the second portion housed two 1-ton cylinders of Cl_2. Selection of the generator was based on specifications outlined by the utility. The generation equipment* operated on a water eductor principle, which produced sufficient vacuum to deliver the 25-percent $NaClO_2$ solution and molecular Cl_2 gas to the reaction column. Both flows were controlled by flow-rate meters, and the system was designed to shut down if the eductor water supply failed or if chemical feed lines broke. The installation at Evansville was capable of producing 14.24 lb/h (6.4 kg/h) of ClO_2, which could be divided in any proportion between the two halves of the treatment facility.

The Evansville utility began using ClO_2 in its treatment scheme in August 1983. The ClO_2 solution produced was divided evenly between the utility's two treatment systems and injected into the raw water influent lines prior to any additional chemical treatment. During the first five months of operation, a variety of ClO_2 dosages was used to determine the resultant THM reduction, to gather data on what percentage of the ClO_2 dosage would appear as total oxidant residuals in the finished water, and to review the general operation of the entire system. During this period, only one major problem was encountered. Brass corporation cocks used to connect the PVC ClO_2 feed lines to the raw water influent piping were oxidized by the concentrated disinfectant. This oxidation and subsequent leaking of ClO_2 solution resulted in disruption of the new treatment technique. The problem was resolved by sliding a section of PVC pipe through new corporation cocks into the main stream of the raw water lines. This modification permitted the PVC piping to serve as an inductor while preventing direct contact of the brass corporation cocks with concentrated ClO_2 solution.

Effect of change to chlorine dioxide. Since the implementation of the treatment change, total oxidant levels have been consistently maintained below the 1.0-mg/L stipulated by the USEPA. With an average ClO_2 dosage of 1.2 mg/L

*Rio Linda Chemical Co. Inc., Sacramento, Calif.

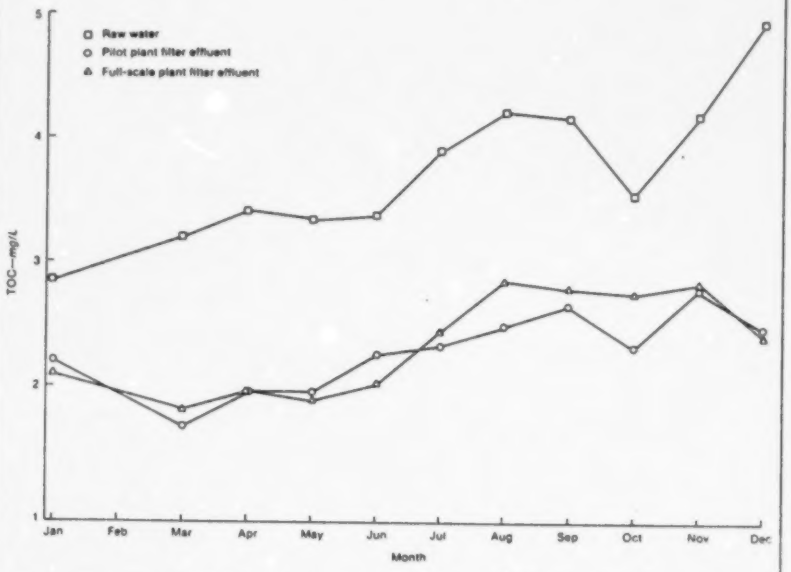

Figure 5. Average TOC concentrations in effluents of full-scale and pilot plants

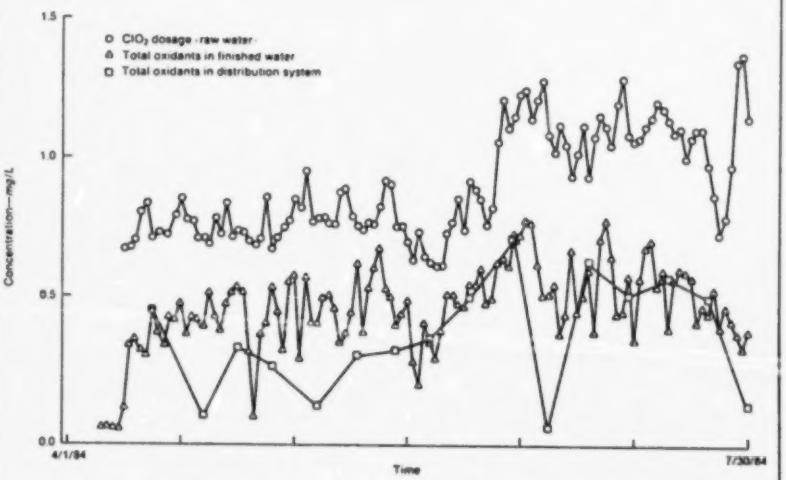

Figure 6. Concentrations of total chlorine dioxide oxidants at various points in Evansville's treatment system

applied during pretreatment, total oxidant concentrations in the finished water have averaged 0.5 mg/L (Figure 6). These data show that approximately 42 percent of the ClO_2 dosage was present as total oxidant in the finished water effluent. The ability to maintain relatively low levels of total oxidants was largely attributable to the generation efficiency of the equipment, with an average efficiency of 95 percent being obtained over an 18-month period. All of the data for total oxidant concentrations and generator efficiency were produced using a recently developed amperometric methodology,[13] which allowed for the determination of ClO_3^- concentrations—a measurement not possible by previously recognized techniques.

A brief study was conducted to determine whether a further reduction in THM formation could be expected by oxidation of precursor material. This reduction would be in addition to that known to be achieved by precursor removal through coagulation and settling prior to Cl_2 addition. Before ClO_2 was added samples from the filter effluents of each of Evansville's two treatment schemes were analyzed for instantaneous THMs. This ensured that any variation in THM concentrations between the two systems would not be attributable to slight differences in their design. These samples indicated a <1-percent difference in the instantaneous THM levels of the two plants (Table 4). With Cl_2 application altered to a point following coagulation and settling in both treatment schemes, an average ClO_2 dosage of 1.4 mg/L was introduced into the raw water line supplying the south plant. During the next three weeks, sample analyses indicated an average reduction in instantaneous THM levels in the south plant of 32 percent (Table 4). This reduction was in comparison with THM levels at the north plant that had already been reduced by altering the chlorination point and is believed to be the result of precursor oxidation.

The bacteriological integrity of the Evansville water supply was never affected by the change in treatment. Although the average ClO_2 dosage did not consistently reduce total coliform and heterotrophic plate counts to acceptable levels, no bacteria were detected after settled water chlorination. A measurable free Cl_2 concentration was maintained throughout the distribution system.

Summary

A pilot-plant study at the Evansville (Ind.) Water and Sewer Utility showed that applying ClO_2 to the raw water could drastically reduce the formation of THMs. Because of this observation and the ability to maintain bacteriological quality without operational problems, ClO_2 was selected for use in the full-scale plant. With the use of ClO_2 as a predisinfectant, the Evansville utility has been able to maintain annual average TTHM levels of between 50 and 80 μg/L in its distribution system.

During the pilot-plant study, total oxidant by-products of ClO_2 in the filter effluent were above the recommended level of 1.0 mg/L. This was caused mainly by the inefficiency of the generator unit. Approval for the use of ClO_2 in the full-scale plant was conditioned on total oxidants being <1.0 mg/L. This level has been maintained without any problems.

With Evansville's ability to adhere to the total oxidant guideline, no adverse health effects are anticipated. Several investigators have evaluated the health effects of ClO_2 and its inorganic by-products, ClO_2^- and ClO_3^-.[14-18] These studies did not indicate any problems at the level recommended for total oxidants.

The use of ClO_2 will increase disinfection costs. Although these costs may seem high, the additional delivered cost to the customer is actually quite low. For instance, in a study by the University of North Carolina,[19] the average water bill increase for a city in South Carolina was found to be $1.77/resident/year.

Acknowledgment

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Mention of trade names or commercial products does not constitute endorsement or recommendation for use by the USEPA.

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